FLARE-NOTE-LAR 56

Argon Purity and Purification in a Large Cryogenic Tank

April 28, 2005

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Abstract

Problems and issues related to the attaining and maintaining high levels of purity of liquid argon in a large commercial cryogenic tank are analyzed. High levels of argon purity can be achieved with modest size recirculation system as th purification may be extended over very long time. Out-gassing from the walls and from the materials inside the tank is of relatively low importance, again because of long periods of time involved. The most important design objective of the experiment is minimization of potential leaks and reduction of their impact on the argon purity by buffer volumes filled with gaseous argon. Gas purification system is found to be of relatively little importance.

1 Introduction

Large Liquid Argon Time Projection Chamber presents an attractive option for a neutrino detector. In addition to its superb particles detection and measurement capabilities a large LAr TPC can be built in a relatively straightforward and economic manner. Principles of the detection technique as well as many of the technical solutions have been established by the ICARUS collaboration.[1].

The fundamental advantage of the LAr TPC technology stems from the possibility of attaining very long (of the order of several meters) drift distances, hence permitting a readout of a very large detector volume with relatively small number of wires and electronics channels. Detector performance is related to two fundamental parameters, signal size, S, and electronics noise, N. The former is intimately related to the purity of argon and in particular to the concentration of electro-negative impurities, oxygen being the principal one.

In this note we examine issues related to the argon purity requirements in FLARE. The relevant features and parameters of the detector are:

- large commercial cryogenic tank
- commercial purification system
- drift distance of 3 m
- wire spacing of 5 mm

2 Requirements

Signal-to-noise ratio, S/N, requirement determines several aspects of the experiment:

- data size after baseline subtraction (if no further signal processing is performed)
- pattern recognition capability
- energy resolution

While S/N of the order of $5-6$ is probably quite sufficient, as shown by the date recorded in T600 module of ICARUS[1] , we feel that the design of a large detector should aim for the S/N of the order of 10, thus to leave some room for possible design optimization. The noise level, in number of electrons, is primarily determined by the detector capacitance. With JFET technology and for the integration time of 1 μs it is of the order

$$N[e] = 500 + 2.6C$$

where C is the detector (i.e. wires and cables) capacitance in pF. FLARE detector is designed is to achieve $N \leqslant 2000\ el$, by reducing the wire capacitance and keeping the cables length below $3-4\ m$.

Minimum ionizing particle produces 55000 e/cm in liquid argon, thus the produced signal as seen by a single wire (with 5 mm spacing) is 27500 electrons. The challenge of argon purification system is to ensure that more than 75% of the produced electrons survive the drift to the wire even for the longest drift path of 3 m.

Drift velocity of electrons in liquid argon depends on the applied electric field. We assume field of 500 V/cm , giving $v_{drift} \approx 1.5\ mm/\mu$sec, thus the longest drift time is 2 ms.

Diffusion coefficient for liquid argon is $D = 4.8\ cm^2/s$, hence the maximal longitudinal spread of the electron signal due to diffusion is

$$\Delta x = \sqrt{2Dt} = 1.4\ mm$$

The primary mechanism of the loss of electrons is the electron attachment to the electro-negative impurities in argon, mostly oxygen. It os a random process which can be parameterized by a characteristic lifetime τ:

$$Q(t) = Q(0)\exp(-t/\tau)$$

A requirement that $Q(2\ ms)/Q(0) \geqslant 0.75$ translates into the requirement that the **electron lifetime τ exceeds** 7 ms.

Electron lifetime is primarily related to the concentration of oxygen in the volume of liquid argon with the approximate relation

$$\tau = 300\ \mu s \frac{O_2\ concentration}{1ppb(O_2)}$$

thus the **required purity of the liquid argon is** 50 $ppt(trillion)$ (oxygen-equivalent).

It should be noted that electron lifetime exceeding 10 ms has been attained using a commercial filter (Oxysorb/Hydrosorb) by ICARUS collaboration in a small test module[2]. In a short operation of a large T600 detector the lifetime of 1.75 ms was attained. Detailed analysis of the argon purity data indicates that the electron lifetime was limited by duration of the run due to out-gassing of the cables ad that it would eventually reach the value of 13 ms[6].

Analysis of the T600 data shows, in particular, that the there were no detectable leaks leading to a degradation of the argon purity.

3 From ICARUS to FLARE

Several measurements indicate that long drift distance can be easily realized with ICARUS-type modules. Whereas it is a good news, it must be stressed

that there are very significant differences between the ICARUS detector and the proposed FLARE detector[3]:

- vessel
  - ICARUS has a custom designed aluminum vessel with thin insulation and active cooling using liquid nitrogen
  - FLARE is using commercial cryogenic tank constructed of 9% nickel steel. Thermal insulation is provided by a thick layer of perlite.
- vessel surface preparation:
  - ICARUS tank walls have been subject to pickling using dilutes solution of nitric acid and to passivation. They were subsequently rinsed with de-mineralized water and dried with dry air
  - FLARE tank will be washed with cleaning solution (no halogens!) and water
- initial conditions
  - ICARUS vessel has been evacuated to 10^{-4} *mbar* before filling with liquid argon
  - FLARE tank will be purged with inert gas (probably argon) before a fill
- leak-tightness
  - all the welds of ICARUS vessel were tested for Helium-tightness by vacuum pumping in the volume of the walls themselves
  - all the welds of the FLARE tank will be tested using automated Ultrasonic Technology. More about leaks in the corresponding section.

In addition to these factors, which may indicate that attaining the ultra-high levels of purity may be considerably more challenging in a large commercial tank and which will be examined in this note, there are several other factors making the large volume easier to purify than the ICARUS detector:

- time factor: continuous purification process monotonically reduces the amount of oxygen in the inner volume (in the absence of leaks). Long fill time, of the order of 9 months, permits removal of a very large amount of oxygen before the start of data taking. This factor also reduces the impact of out-gassing.
- surface-to-volume ratio. The contaminating oxygen comes primarily from the surfaces tank walls and the readout cables. These surfaces grow like a square of linear dimensions, hence their effect on the argon purity diminishes inversely with the linear dimensions of the tank

- emergence of several industries, chiefly the silicon chip manufacturing, utilizing ultra-pure argon has stimulated development of the industrial purification methods. There are several new filtering agents, beyond the one used by ICARUS, and, in particular, there are off-the-shelf units for large scale purification of liquid argon.

4 Argon Purity in the Tank: The Model

Modeling of the argon purity level in a large tank is relatively straightforward. Owing to chemical inertness of argon and cryogenic temperatures there are no chemical reactions taking place, hence there is no sources of oxygen.

Tank consists of two vessels: an inner tank and the containment vessel. The space between them is filled with perlite and purged with dry argon. Tank interior contains two volumes: that of the liquid argon (LAR) and the argon gas (GAR) at the top of the tank. There two penetrations of the inner tank walls:

- inlet pipe reaching to the bottom of the tank and used for the initial fill
- a liquid argon purification loop: an outlet pipe near the bottom of the tank and a return pipe

Re-liquefaction of the boil-off argon is accomplished with liquid nitrogen condensers at the top of the tank: gaseous argon re-condenses and rains back to the liquid volume without leaving the tank. Liquid nitrogen pipes do not communicate with the tank volume. In principle, one can consider another purification loop, that of the gaseous argon. Its usefulness is very limited in the FLARE tank geometry due to the fact that liquid argon volume absorbs the oxygen from the gas volume. In equilibrium oxygen-to-argon ratio is the same, per unit mass, in the gas and liquid phase[5], hence the oxygen concentration per unit volume is som 300 times higher in the liquid phase. In a tank with external cooling this leads to a saturated layer of liquid argon with diffusion as a means of transporting the oxygen into the interior of a tank. In FLARE configuration the convective currents transport the oxygen to the inner volume and to provides very thorough mixing of the liquid[4].

4.1 The Tank

We assume that the tank holding liquid argon has 40 meter diameter and 30 meter height. 50 kton of liquid argon will fill this tank up to the height of 28.4 meters. The reminder of the volume, ullage, will contain support structures (trusses), wire mounting boards and cables.

4.2 Oxygen Balance in the Gas Volume

Change of the oxygen content in the GAR volume can be written as

$$\frac{dM_{O_2}^G}{dt} = OG_{cable}S_{cables} + OG_{walls}S_{walls} + Leak - T_{G\_to\_L}$$

where:

- $M_{O_2}^G$ - mass of oxygen in th gas volume (above the liquid phase)
- OG_{cable} - out-gassing rate of the signal cables. It is usually expressed in terms $TorrLiters/(cm^2s)$ or equivalent units. In the following we will use a typical value for polyolefine cables of $1 \times 10^{-6} TorrLiters/(cm^2s)$. Out-gassing rate diminishes with time like $1/t$ until the total amount of oxygen at the surface is exhausted and it converges to the $1/\sqrt{t}$ behavior characteristic for diffusion process.
- S_{cables} - total area of the signal cables. We will use 1200 m^2 as an estimate of this area.
- OG_{walls} - out-gassing rate of the tank walls. It is expressed in the same units as the out-gassing rate of cables. It has been shown that the only 'warm' vessel surfaces, in contact with the gas phase, contribute to the out-gassing process. 'Cold' surfaces submerged in the liquid act as oxygen getters and contribute to the purification process. We will ignore this contribution in the following for two reasons: first it may depend on the treatment of the vessel surfaces and second the purification action may saturate once the tank walls are covered with a mono-layer of oxygen. Out-gassing coefficient of steel varies greatly, in the range $7 \times 10^{-10} - 7 \times 10^{-7} TorrLiters/(cm^2s)$[7], depending on the grade of the material and the surface preparation. In the following we will assume that there is no special treatment of the tank walls and that the out-gassing rate of the walls is the same as the plastic cables.
- S_{walls} - total area of 'warm' tank surfaces.This area diminishes linearly with time during the filling process. The final value, relevant for the running of the experiment, depends somewhat on the height if th gaseous layer on top of the liquid volume and on the support structure for the wire planes. In the following we will use $S_{walls} = 1700\ m^2$ during the operation phase of the detector.
- $Leak$ - rate the air entering the gas volume, in cm^3/s, due to possible imperfection of the tank walls and/or piping system.
- $T_{G\_to\_L}$ - rate with which the oxygen is transferred from the gas to the liquid volume. It is calculated as a product of oxygen concentration in liquid (up to 11% correction due to differences between the concentrations

in gas and liquid phases) and a rate of the downward mass transfer near the surface of the tank. If calculated amount of oxygen transfered to liquid volume in a given amount of time exceeds the total amount of oxygen in the gas phase, the latter is used instead.

As mentioned before a possible reduction of the oxygen content in the gas phase with a dedicated gas purification system is neglected. Its inclusion in the model with GAR recirculation rate of 1000 $m^3/hour$ changes (improves) the level of purity in LAR by $10-20\%$. This improvement is realized, however, only at very late times when the argon purity is already at very high levels. Gas phase purification is effective in removing the oxygen out-gassed from the materials and their contribution to the oxygen level in the liquid is likely to be much smaller than the contribution of the input material during the fill time.

4.3 Oxygen Balance in the Liquid Volume

Change of the oxygen content in the liquid volume can be written as

$$\frac{dM^L_{O_2}}{dt} = T_{G\_to\_L} - Rate_{recirc}\frac{M^L_{O_2}}{M_{Ar}}\varepsilon_{filter}$$

where:

- $M^L_{O_2}$ - mass of oxygen in the liquid volume
- $T_{G\_to\_L}$ - rate with which the oxygen is transferred from the gas to the liquid volume. It is calculated as a product of oxygen concentration in liquid (up to 11% correction due to differences between the concentrations in gas and liquid phases) and a rate of the downward mass transfer near the surface of the tank. If calculated amount of oxygen transfered to liquid volume in a given amount of time exceeds the total amount of oxygen in the gas phase, the latter is used instead.
- $Rate_{recirc}$ - rate of recirculation of liquid argon through the purification system
- M_{Ar} - total mass of liquid argon
- ε_{filter} - efficiency of the filter defined as a ratio of output-to-input oxygen levels. This ratio is typically of the order of 10^{-4} for the industrial filters. This performance is achieved for the input purity of liquid argon at the level of 10^{-6}. It is very likely that the filter performance drops significantly for much higher purity of the input. We have assumed here $\varepsilon_{filter} = 0.01$. It is not a critical parameter as long as it is in the range below $0.1-0.2$.It enters the balance equation as a factor multiplying the flow rate, hence in practical terms the purification power of the filter determines the effective flow of rate.

4.4 Fill Procedure

There are several possible models of the fill procedure. As an example we use the following scenario:

- liquid argon is received at the rate of 8 tons/hour (about 200 tons per day)
- initial argon quality is evaluated and 'bad' batches are rejected.
- argon is purified to the level below 10^{-10} at the average rate of 8 tons/hour (5700 liters/hour) and stored in the intermediate storage tanks
- ultra-pure argon is transferred to the main detector tank. We allow for the possibility that the transfer lines may contaminate the ultra-pure argon the purity of the argon transferred to the main tank may be worse than that in the intermediate tanks. Even if the purity of the argon cannot be maintained at 10^{-10} level during transfer the initial purification is an important step to ensure that the argon is purifiable to the required levels.

Fig.4.4 shows the evolution of relevant parameters: mass of liquid argon, surface of 'warm' surfaces contribution to the out-gassing and the size of the gas volume during the fill procedure.

4.5 Main Tank Purification System

We assume that the main tank will be equipped with a recirculation system as shown in Fig.4.5.

It consists of a recirculation loop pumping the liquid argon through a purifier and returning the purified argon to the main tank. For the analysis of the argon purity the important parameters of the purification system are the total recirculation rate, ton/hour, and the efficiency of the purification system. The latter is not known very well for the input argon purities at the level of 10^{-11} and it is assumed to be $\varepsilon = 0.01$, some factor 100 worse that the performance of industrial filters at the input purity of the order of 10^{-6}.

5 Evolution of Argon Purity in the Main Tank

We examine the evolution of the argon purity in several steps. We start with the simplest case of filling a perfectly pure tank with no sources of internal contaminants. In the next step we introduce the out-gassing of materials in the tank. Large cryogenic tanks cannot be evacuated, hence they do contain large amount of oxygen at the very beginning of the fill procedure. The problem of the initial purge is considered next. Long term operation of the experiment require that the design level of argon purity is maintained over long period of time ad that adequate mitigation methods are available to deal with even most improbable mishaps.

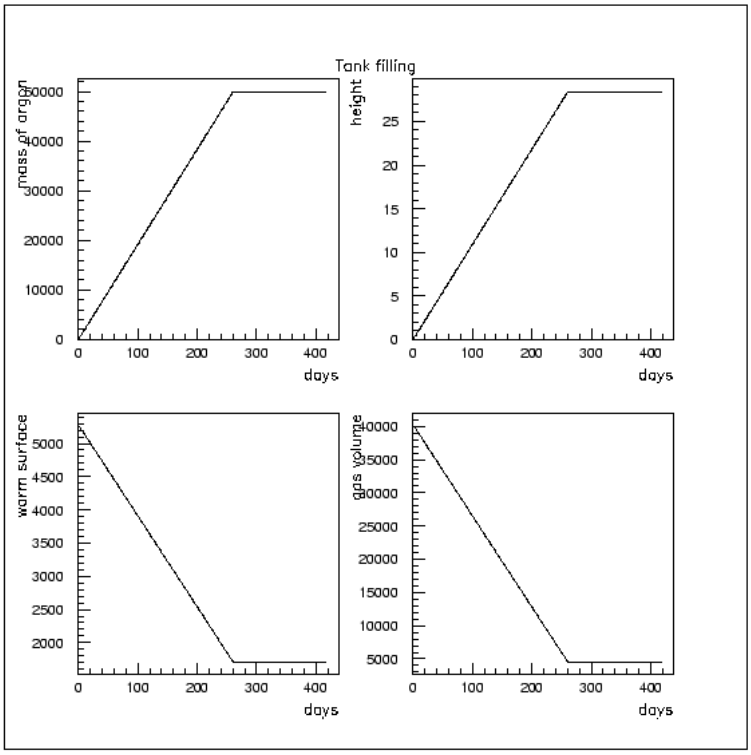

Figure 1: Filling process: evolution of the mass of liquid argon, height of the liquid in the tank, warm tank surfaces and the gas volume with time.

5.1 Filling a Pure Tank

Fig.5.1 shows the purity of argon in the main tank as a function of time during and after the fill process for different levels of purity of the incoming liquid argon. The assumed recirculation-purification rate 100 $tons/hour$.

In this simplified case the purity of liquid in a tank during the fill process is determined by the equilibrium between the incoming amount of oxygen and the oxygen removed by the purification system. Th exact level of this equilibrium depends on the purification rate, in the considered case of 100 $tons/hour$ it corresponds to about 8% of the input argon purity. After the fill is completed argon purity improves with time like e^{-N_V} where N_V is the number of exchanged (purified) volumes and it depends on the recirculation rate.

$$N_V(t) = \frac{Rate_{recirc}}{100\ tons/hour} \frac{t}{20.8\ days}$$

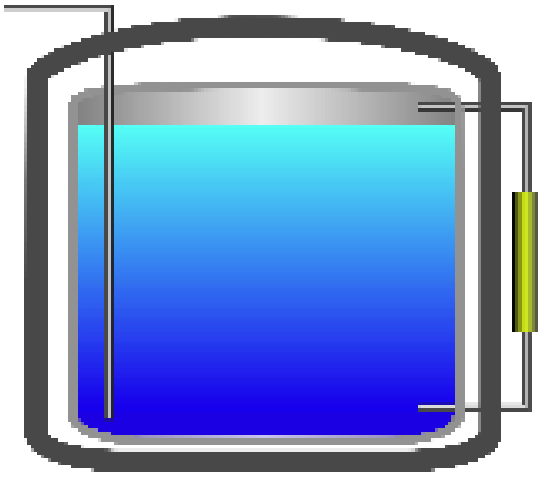

Figure 2: Liquid argon lines for the main detector tank: the filling lines on the left and the recirculation/purification lines on the right.

5.2 Out-gassing from Materials inside the Tank

All warm surfaces inside the tank will be sources of out-gassing. The out-gassing rate depends on the temperature of the surface, it is expected to be much smaller at the cryogenic temperatures than the rate measured at room temperatures. Out-gassing rate slows down with time approximately like $1/t$. The exact rate of out-gassing depends on the surface treatment and on the history of the material, hence the existing data may appear to be inconsistent, at times. Analysis of the out-gassed materials shows that it is dominated by water, hydrogen, CO and CH_4.Composition of the out-gassed materials changes with time, slowly. Typically, oxygen constitutes about 1% of the out-gassed materials.

In the following we will assume that the tank contains 1200 m^2 of ribbon cables and that they outgass with the initial rate of 1.0^{-6} $TorrLiter/cm^2$ sec. We do not assume that the tank walls will not undergo a special treatment, hence we assume that the warn tank surfaces outgass with th same rate as plastic cables.

To set the scale for the impact of out-gassing on argon purity we notice that after 10 days of the filling procedure the amount of oxygen out-gassed from the cables and tank walls is of the order of 0.2 g/day and it is the same as the amount of oxygen introduced into the tank volume with new argon if its purity is 1 *ppb*. If the purity of newly introduced argon can be maintained at th level

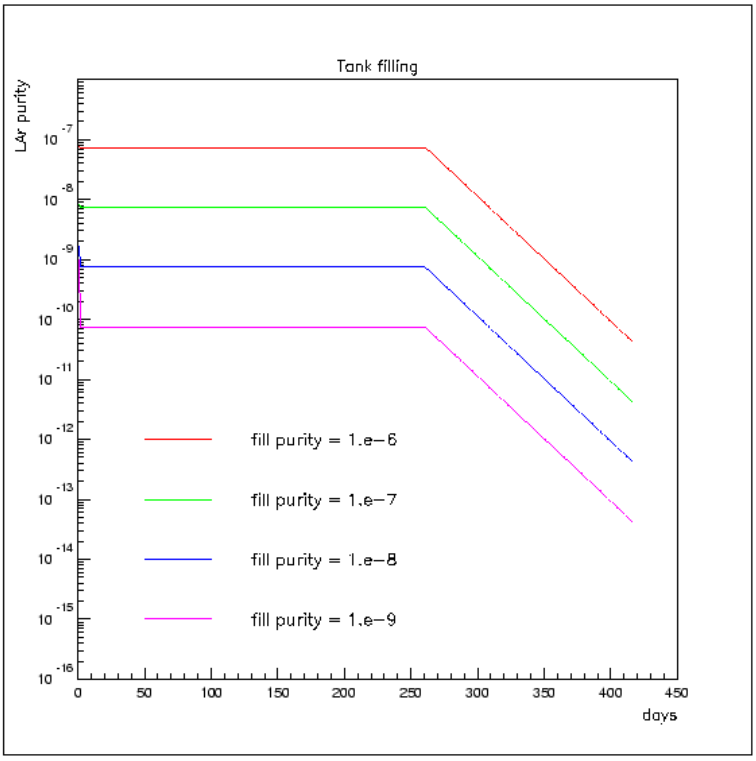

Figure 3: Purity of liquid argon inside the tank as a function of time during and after the fill for different purities of input argon. 100 *tons/hour* purification system is assumed.

of 0.1 *ppb* then the out-gassing of walls and cables dominates for the first 100 days of the fill procedure. These consideration indicate that the argon purity in the main tank will be primarily determined by the purity of argon and that there are not much incentives to go to extraordinary efforts in cleaning the tank walls.

Fig.5.2 shows the argon purity levels which can be attained with 28 $t/hour$ recirculation system for two plausible filling scenarios: 1 *ppb* and 0.1 *ppb* input argon purities. In the latter case the design argon purity is achieved already at the end of the fill stage whereas the former one requires additional two month of continuous purification. This example indicates the importance of maintaining argon purity during the transfer from the intermediate storage tanks to the main tank.

To evaluate the contribution of out-gassed materials to the argon purity we have considered a case that the purification system is turned off after 600 days since the beginning of the fill. The out-gassing rate at such late times is expected

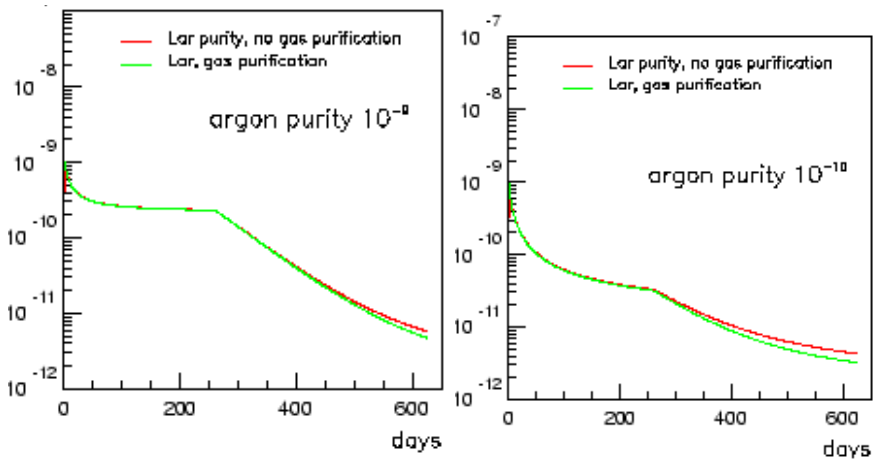

Figure 4: Argon purity in the tank as a function of time for two scenarios of the fill. Red curve is for the case of liquid purification only, green curve is with additional gas phase purification. Out-gassing contributes visibly to the impurities at the beginning of the fill process, especially if the fill is performed with very pure argon.

to be very low, thus allowing operation of the experiment for extended periods of time without the purification system, as shown in Fig.5.2.

5.3 The Initial Purge

Large cryogenic tanks cannot be evacuated, hence the initial atmosphere, some 40 tons of nitrogen and 10 tons of oxygen must be removed by purging. Three volume exchanges can reduce these amounts to 500 kg and 2 tons, respectively. The reduction of the oxygen and nitrogen content may be even greater if the purge argon is introduced at the bottom of the tank (as a liquid, perhaps). The resulting stratification of the atmosphere, owing to the differences in densities, may lead to increased venting of oxygen and nitrogen. Purging process with argon can be continued, but the removed amount of oxygen and nitrogen will be reduced progressively.

A possible scheme of removal of the rest of the initial atmosphere uses the fact that in the equilibrium state the concentration of oxygen in the gaseous and liquid argon is nearly the same. Given the fact that densities of these wo phases differ by a factor of 300, most of the oxygen is transfered to the liquid phase: liquid acts as a sponge. To ensure the equilibrium some proper mixing mechanism may be necessary.

A very effective method of purification of the tank atmosphere after the initial few (3?) volume exchanges consists of deposition of some 100 tons of liquid argon in the tank and its continuous purifica-

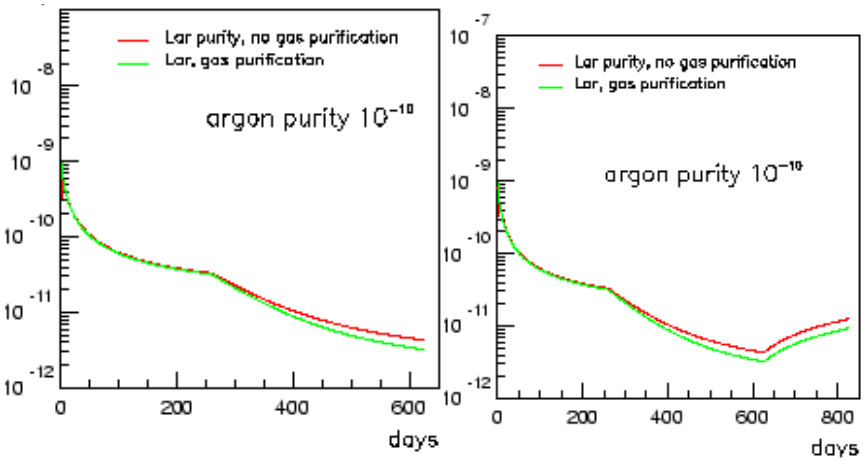

Figure 5: Evolution of argon purity in the tank with 28 *tons/hour* per hour purification and filled with argon of purity 10^{-10} (left) and the same for the case with the purification system stopped after 600 days(right).

tion system. A 28 *tons/hour* purification system would enable several volume exchanges per day, thus permitting to achieve ultra-high levels of purity within several days. This procedure will be a important milestone demonstrating the perform ace of the purification system. The fill procedure will be continued after this initial milestone is reached.

5.4 Leaks

Above examples illustrate the fact that a continuous recirculation/purification of liquid argon will allow to overcome even major amount of oxygen in the main tank. The time to attain desired level of purity depends on the purification rat. The key to this is the fact that the amount of oxygen inside the tank is finite, hence continuous purification will reduce it to the negligible level after some amount of time.

This is not true, naturally, if there are leaks in the system. The detailed analysis of the effect of leaks is difficult, as the tank volume is under positive pressure, hence reducing any back-flow of air into the tank. It depends, naturally, on the size of the leak. Fig.5.4 illustrates the case where there is an effective intake of air into the tank at the rate of 0.001 $cm^3/\,\mathrm{sec}$. The effect of leaks is similar to the filling process: there is a constant rate of oxygen introduced to the tank. In such a case the liquid purity reaches a plateau at the level dependent on the purification rate.

It is quite clear that a sizable leak may lead to a degradation of argon purity beyond acceptable levels, hence that tank construction must include very thorough quality control phase. To ensure long time reliable operation of the

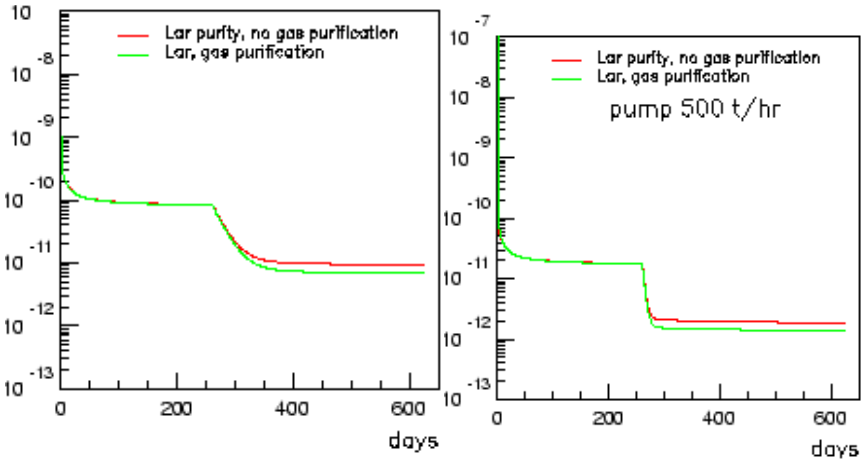

Figure 6: Evolution of the argon of purity in the presence of a leak introducing 0.001 cm^3/sec of air into the tank volume for different sizes of the recirculation system: 100 $tons/hour$ (left) ad 500 $tons/hour$(right).

experiment it is important to reduce or even eliminate the negative impact of leaks. This can be achieved relatively easily by ensuring that all volumes adjacent to the tank and all the transfer lines are either evacuated or purged with argon.

6 Acknowledgments

Many people have contributed to the understanding and the analysis presented in this note. We would like at express our thanks in particular to Francesco Arneodo, Flavio Cavanna, Cary Kendziora, Claudio Montanari, Francesco Pietropaolo and Rich Schmitt.

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